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(54) Title: EPOXY RESINS COMPATIBLE WITH POLYAMINE CURING AGENTS

(57) Abstract

The incorporation of polyurethane poly(meth)acrylate(s) into a composition comprising epoxy resin(s), multifunctional (meth)acrylate ester(s) of a polyol which forms an ester with (meth)acrylic acid, and a polyamine curing agent results in cured epoxy resin(s) that are hard and nonbrittle with high impact resistance and finds use as an adhesive or coating.

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EPOXY RESINS COMPATIBLE WITH POLYAMINE CURING AGENTS

BACKGROUND OF THE INVENTION

1. Field of the Invention

This invention relates to epoxy resins and their uses. More particularly, the invention relates to epoxy resin compositions compatible with polyamine curing agents.

2. Statement of Related Art

U.S. 4,051,195 relates to blends of an epoxide resin and a poly(meth)acrylate ester, which cure rapidly with aliphatic polyamine curing agents even at low temperatures. Such compositions can be employed as coatings, adhesives, castings, moldings and the like at high solids contents, including 100% solids content (so-called solventless compositions).

While these compositions comprise an important advance in the art, since the cured compositions can be formulated to range from flexible to very hard, the latter type tend to be brittle and to have low impact resistance, which are serious disadvantages for their use as coatings, moldings, castings, and the like.

U.S. 4,547,562 discloses solventless polymeric compositions suitable for coatings and moldings formed by the addition reaction product of a first component comprising mono, di or poly amines, amine-terminated structures or an adduct of mono, di or polyamines with mono, di or polyfunctional acrylates or epoxides; and the second component comprising mono, di or poly acrylates, mixtures of mono, di or poly acrylates, or a mixture of mono, di or poly acrylates

and mono, di or polyfunctional epoxides and/or glycidyl esters of acrylic acid or methacrylic acid.

Such compositions when cured can be used as adhesives, varnishes, lacquers, paints, or sealants. However, here again when used as coatings the coatings tend to be brittle, with low impact resistance.

This patent also contains the teaching that isocyanate and hydroxyl-terminated polyurethanes have high viscosities and "cannot be used as components of a liquid, solventless system" (column 3, lines 1-8).

DESCRIPTION OF THE INVENTION

Other than in the operating examples, or where otherwise indicated, all numbers expressing quantities of ingredients or reaction conditions used herein are to be understood as modified in all instances by the term "about".

It has now been discovered that the presence of small quantities of polyurethane poly(meth)acrylates in certain two component liquid solventless epoxy resin systems results in cured epoxy resins that are hard, nonbrittle, and possess high impact resistance. This result is surprising, especially in view of the teachings of U.S. 4,547,562 that hydroxyl-terminated polyurethanes cannot be used in such systems.

The two component epoxy resin system of the invention comprises

A) a liquid epoxy resin component containing

   a) at least one epoxy resin having at least two 1,2-epoxy groups;

   b) at least one multifunctional (meth)acrylate ester which is a polyol having at least two hydroxyl groups, in the form of an ester with acrylic acid or methacrylic acid; and

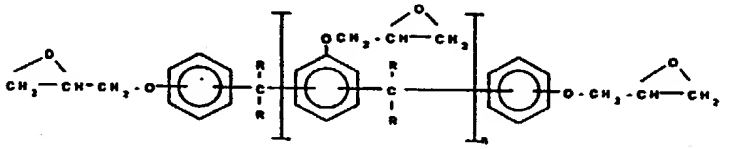
c) at least one polyurethane poly(meth)acrylate;

and

B) a liquid hardener component containing a polyamine curing agent.

The epoxy resin (component A) a) above) includes those epoxy resins having two or more 1,2-epoxy groups per molecule. The epoxy resins include those set forth in U.S. 4,051,195, which patent is expressly incorporated herein by reference. Such epoxy resins are liquid, saturated or unsaturated, aliphatic, cycloaliphatic or heterocyclic and can be either monomeric or polymeric.

Preferred epoxy resins for use herein are the epoxy novolac resins of formula I



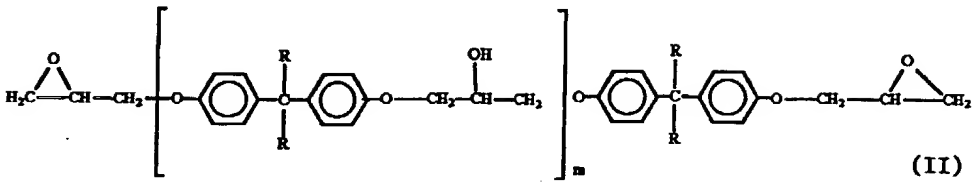
(I)

where R = H or CH_3 and

n = 0 to 4.

The above epoxy novolac resins of formula I are usually mixtures of ortho, meta and para isomers.

Also preferred are epoxy resins of formula II



(II)

wherein R= H or CH_3 and

m = 0 to 4.

Even more preferred are epoxy resins of formula II which are the diglycidyl ethers from epichlorohydrin and bisphenol A or bisphenol F, i.e. compounds of Formula II where R is methyl or hydrogen and m = 0.

The multifunctional (meth)acrylate esters (component A)b) above) useful in the practice of the invention include those esters set forth in U.S. 4,051,195, which is expressly incorporated herein by reference. These multifunctional

(meth) acrylate esters should have a viscosity not greater than 5000 cps, and preferably not greater than 500 cps. Preferred esters are diols or triols, optionally ethoxylated and/or propoxylated with from 1 to 8 EO and/or PO groups, wherein all hydroxyl groups are in the form of acrylate esters, e.g. trimethanol triacrylate. Most preferred are esters of the following formulae:

$$CH_2 = CH - COO(CH_2)_xOOC - CH = CH_2 \quad (III)$$

wherein x is an integer of from 2-6, and

$$CH_2 = \underset{\displaystyle R^1}{\underset{|}{C}}H - COO(CH_2CHO)_yOC - CH = CH_2 \quad (IV)$$

wherein y is an integer of from 1-6 and R^1 is H or CH_3.

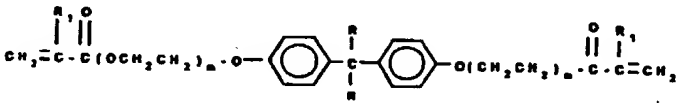
Examples of compounds within the above formulae include 1,3-propanediol diacrylate, 1,6-hexanediol diacrylate, and tripropyleneglycol diacrylate. Where multifunctional (meth) acrylate esters having a viscosity above 500 cps are present, they are preferably present in relatively small quantities, e.g. from 0.1 to 25%, based on the total weight of (meth)acrylate ester. Such higher viscosity esters are e.g. sugar polyacrylates, and in fact their presence may be useful in increasing the crosslink density of the cured epoxy.

While multifunctional methacrylate esters are within the scope of the invention, these esters are usually somewhat less desirable for use herein than the corresponding multifunctional acrylate esters due to their somewhat slower rates of reaction and more limited miscibility with the epoxy resins.

Where less than all hydroxy groups in the multifunctional (meth)acrylate esters are in the form of esters of (meth)acrylic acid, e.g. in the case of a triol or higher polyol having one or more free hydroxyl groups, the nonesterified

hydroxyl groups can be present as such or in the form of C_1-C_3 alkoxy groups, preferably methoxy. It is however usually preferred to esterify all hydroxyl groups with (meth)acrylic acid.

It has also been discovered, and is a feature of this invention, that di(meth)acrylate esters of the formula



(V)

wherein R is H or CH_3,

R_1 is H or CH_3,

n is an integer of from 0-6,

m is an integer of from 0-6, and

n + m = 1-6, preferably 2-6,

when used as component A)b) above, greatly improves the impact resistance and flexibility of the cured compositions, even without the presence of any polyurethane di(meth)acrylate (component A)c) above).

The polyurethane poly(meth) acrylates (component A)c) above) contain from 2 to 6 (meth) acrylate groups, are aliphatic or aromatic polyurethane derivatives, are highly viscous or solid, having a weight average molecular weight in the range of from 1200 to 5000, preferably 1200 to 1800. They are employed in amounts ranging from 0.1 to 25%, preferably 1 to 15%,and more preferably 1 to 10% by weight, based on the combined weight of components A)a) and A)b). Preferred are polyurethane di-acrylates having a weight average molecular weight in the range of from 1200 to 1800. These polyurethane diacrylates are usually prepared by reacting a diisocyanate with hydroxyethyl acrylate in a 1:1 molar ratio, so that the resulting adduct contains a free isocyanate group, and then reacting two moles of this adduct with one mole of a dihydroxy polyether or polyester.

The hardener component (component B)) of the present two component epoxy resin systems are aliphatic polyamines such as polyalkylene polyamines, e.g. ethylenediamine ($H_2NCH_2CH_2NH_2$) dimethylethylene diamine ($CH_3NHCH_2CH_2NHCH_3$), diethylenetriamine ($H_2NCH_2CH_2NHCH_2CH_2NH_2$), tetramethylene diamine ($H_2NCH_2CH_2CH_2CH_2NH_2$), triethylenetetramine ($H_2NCH_2CH_2NHCH_2CH_2NHCH_2CH_2NH_2$), hexamethylene diamine

($H_2N(CH_2)_6NH_2$), etc. Preferred for use herein are polyamide curing agents, i.e. amidoamines obtained by reacting a polyamine such as the above with straight or branched chain saturated aliphatic polycarboxylic acids, e.g. having from 2-50 carbon atoms in the alkylene groups, in which each carboxylic acid group has been reacted with a molecule of a polyamine. The resulting polyamides must contain at least two amine groups that are primary and/or secondary in nature. Preferred polyamide curing agents are the reaction products of one mol of a C_{36} fatty diacid from the dimerization of a C_{18} unsaturated fatty acid with two mols of diethylenetriamine or triethylenetetramine. Also, polyamines containing epoxy groups can be employed herein.

In addition to the polyamine curing agent, the hardener component can optionally contain quantities of other ingredients, e.g. from 0.001 to 65% by weight, based on the weight of polyamine curing agent, of ingredients such as accelerators, fillers, reinforcing agents, dyes, pigments and other additives.

In the compositions of the invention, the percentage ratio by weight of component A)a) to component A)b) is from 85:15 to 20:80. The particular ratio selected within the above range is not critical and will depend on the particular components chosen and the properties desired for the cured epoxy product.

Components A and B are used in an equivalent ratio of epoxy groups plus acrylate groups in the epoxy resin component to amine hydrogen atoms in the hardener component of from 0.85:1 to 1:1.15, preferably from 0.95:1 to 1:1.05, and more preferably 1:1.

The two component epoxy resin systems of the invention are applied by mixing together the epoxy resin component and the hardener component in the

absence of solvents and applying the mixture at a temperature of from 0° to 150°F, preferably from 40° to 110°F, and more preferably from 40° to 95°F.

It is of course also possible to add solvents thereto, such as hydrocarbons, ethers, alcohols, esters, ketones, etc., but due to the liquid nature of the present compositions, such solvents are unnecessary. Moreover, for many applications solvents are highly undesirable, acting to reduce the solids contents, presenting environmental and safety problems, and causing a reduction in physical properties, e.g. in thick coatings of 20-1000 mils, solvents will be trapped in the coatings, resulting in cheesey films.

The two component epoxy resin system can be used as an adhesive system to bond two like or different substrates together as with other epoxy adhesives; as tough resilient nonbrittle coatings having high impact resistance for hard surfaces, including coatings for floors; as casting, molding and potting compositions; and as encapsulating compositions.

The epoxy system can be used immediately after mixing due to its low viscosity, even at a relatively high solids content, and its increased reactivity even at relatively low cure temperatures. Short cure times are also achieved. The cured mixtures exhibit excellent flexibility, impact resistance, and good chemical resistance.

Since the present epoxy system does not require any organic solvent, thick coatings containing 100% solids content, e.g. from 0.5 to 1 inch thick, can be readily applied to a substrate.

The epoxy systems of the invention can also be used as paint compositions, when mixed with paint pigments. The paint coatings resulting from

their use are unusually tough, adhesive and chemically resistant.

The invention will be illustrated but not limited by the following examples.

EXAMPLES

Example 1

A two component epoxy resin system is prepared from the following ingredients:

COMPONENT A:

Ingredient	parts per hundred	acrylate equiv. wt.	epoxy equivalents
DER 331[1]	60	190	0.316
PHOTOMER® 6022[2]	20	130	0.154
PHOTOMER® 4127[3]	20	180	0.111
	100		0.581

COMPONENT B:

Ingredient	parts per hundred parts of Component A	amine equiv. wt.	amine hydrogen equivalents
DETA[4]	12	20.6	0.581

[1]DER 331 is the diglycidyl ether of bisphenol A.

[2]PHOTOMER® 6022 is an aromatic polyurethane containing six acrylate groups, a viscosity of from 500-800 poises, and a weight average molecular weight of 800.

[3]PHOTOMER® 4127 is a neopentylglycol- 2PO- diacrylate.

[4]DETA is diethylenetriamine.

Example 2

A two component epoxy resin system is prepared from the following

ingredients:

COMPONENT A:

Ingredient	parts per hundred	acrylate equiv. wt.	epoxy equivalents
DER 331	60	190	0.316
PHOTOMER® 6022	20	130	0.154
PHOTOMER® 4127	20	180	0.111
	100		0.581

COMPONENT B:

Ingredient	parts per hundred parts of Component A	amine equiv. wt.	amine hydrogen equivalents
VERSAMINE® A-50[5]	25	43	0.581

[5]VERSAMINE ® A-50 is an epoxy derivative of diethylenetriamine.

Example 3

A two component epoxy resin system is prepared from the following ingredients:

COMPONENT A:

Ingredient	parts per hundred	acrylate equiv. wt.	epoxy equivalents
DER 331	75	190	0.395
PHOTOMER® 4025[6]	25	270	0.093
	100		0.488

COMPONENT B:

Ingredient	parts per hundred parts of Component A	amine equiv. wt.	amine hydrogen equivalents
DETA	10	20.6	0.488

[6]PHOTOMER® 4025 is bisphenol A-8E0- diacrylate.

Example 4

A two component epoxy resin system is prepared from the following ingredients:

COMPONENT A:

Ingredient	parts per hundred	acrylate equiv. wt.	epoxy equivalents
DER 331	75	190	0.395
PHOTOMER® 4025[6]	25	270	0.093
	100		0.488

COMPONENT B:

Ingredient	parts per hundred parts of Component A	amine equiv. wt.	amine hydrogen equivalents
VERSAMINE ® A-50	21	43	0.488

CLAIMS

1. A two component epoxy resin system comprising

A) a liquid epoxy resin component containing

a) at least one epoxy resin having at least 2 1,2-epoxy groups;

b) at least one multifunctional (meth)acrylate ester which is a polyol having at least two hydroxyl groups in the form of an ester with acrylic acid or methacrylic acid; and

c) from about 0.1 to about 25% parts by weight, based on the weight of a) plus b), of at least one polyurethane poly(meth)- acrylate; and

B) a liquid hardener component containing a polyamine curing agent.

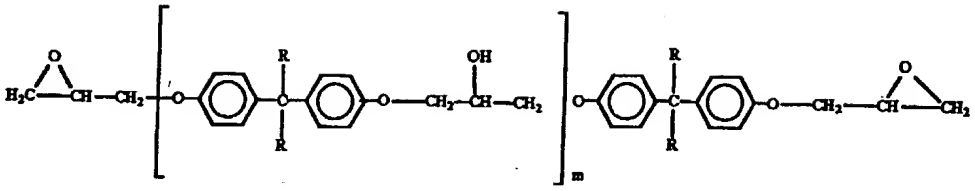
2. The epoxy resin system of claim 1 wherein from about 1 to about 15% by weight of component A)c) is present in the epoxy resin component.

3. The epoxy resin system of claim 1 wherein component A)c) has a weight average molecular weight in the range of from about 1200 to about 5000.

4. The epoxy resin system of claim 3 wherein the molecular weight of component A)c) is in the range of from about 1200 to about 1800.

5. The epoxy resin system of claim 1 wherein the percentage ratio by weight of component A)a) to component A)b) is from about 85:15 to about 20:80.

6. The epoxy resin system of claim 1 wherein component A)a) is an epoxy novolac resin of formula II



(II)

wherein R = H or CH_3, and n = 0 to 4.

7. The epoxy resin system of claim 6 wherein n in formula I is from 0-2.

8. The epoxy resin system of claim 6 wherein n in formula I is equal to zero or 1.

9. The epoxy system of claim 1 wherein component A)b) has a viscosity not greater than about 5000 cps.

10. The epoxy system of claim 9 wherein said viscosity is not greater than about 500 cps.

11. The epoxy system of claim 1 wherein component A)b) is at least one of the following:

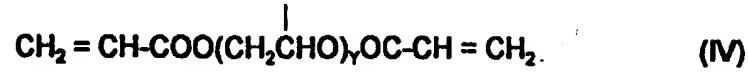
(i) a diol or triol having all hydroxy groups in the form of an acrylate ester and wherein the diol or triol is optionally ethoxylated and/or propoxylated with from 1 to 8 EO and/or PO groups;

(ii) an acrylate ester of the formula $CH_2 = CH - COO(CH_2)_xOOC - CH$

$=CH_2$ (III)

wherein x is an integer of from 2-6; and

(iii) an acrylate ester of the formula



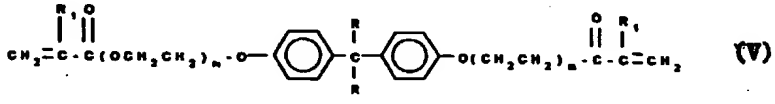
$CH_2 = CH\text{-}COO(CH_2\underset{\displaystyle R^1}{\underset{|}{C}}HO)_YOC\text{-}CH = CH_2$ (IV)

wherein y is an integer of from 1-6 and R is H or CH_3.

12. The epoxy resin system of claim 1 wherein the polyamine curing agent of component B) is a straight or branched chain saturated aliphatic polycarboxylic acid in which each carboxylic acid group is in the form of an amide with a polyamine, wherein the amide contains at least two primary and/or secondary amine groups.

13. The epoxy system of claim 1 wherein component A)b) is at least one di(meth)acrylate ester of the formula

this is revised



(V)

wherein R is H or CH_3,

R_1 is H or CH_3,

n is an integer of from 0-6,

m is an integer of from 0-6,

and n + m = 1-6.

14. The epoxy system of claim 13 wherein in formula IV n + m = 2-6.

15. A curable liquid epoxy resin component comprising

a) at least one epoxy resin containing at least two 1,2-epoxy groups;

b) at least one multifunctional (meth)acrylate ester which is a polyol having at least two hydroxyl groups in the form of an ester with acrylic acid or methacrylic acid; and

c) from about 0.1 to about 25% parts by weight, based on the weight of a) plus b), of at least one polyurethane poly(meth)acrylate.

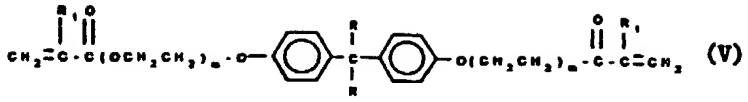
16. The epoxy resin component of claim 15 wherein the percentage ratio by weight of component A)a) to component A)b) is from about 85:15 to about 20:80, and from about 1 to about 15% by weight of component A)c) is present therein.

17. A two component epoxy resin system comprising

A) a liquid epoxy resin component containing

a) at least one epoxy resin having at least two 1,2-epoxy groups, and

b) at least one di(meth)acrylate ester of the formula



(V)

wherein R is H or CH_3,

R_1 is H or CH_3,

n is an integer of from 0-6,

m is an integer of from 0-6,

and n + m = 1-6;

and

B) a liquid hardener component containing a polyamine curing agent.

18. The epoxy resin system of claim 17 wherein in component A)b) n + m = 2-6.

INTERNATIONAL SEARCH REPORT

Int.. .tional application No.
PCT/US94/14550

A. CLASSIFICATION OF SUBJECT MATTER
IPC(6) : C08L 63/02, 63/04
US CL :525/502, 532
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
U.S. : 525/502, 532

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
USPTO APS AND ORBIT WPAT (poly)urethane (poly)(meth)acrylate and epoxy)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US, A, 5,198,524 (BUSH ET AL.) 30 March 1990, col. 5, lines 32, 48 and 49.	1-18
Y	US, A, 4,755,571 (IRVING ET AL.) 05 July 1988, see cols. 5-6, formula V; col. 6, lines 59-60; col. 9, lines 52-54; col. 11, Epoxide II and Acrylic ester III and col. 12, Example 2.	1-18
Y	JP, A, 4-149443 (MITSUBISHI ELECTRIC CORP) 22 May 1992, Derwent Accession No. 92-223594/27, see lines 12-22.	1-18
Y	JP, A, 4-149444 (MITSUBISHI ELECTRIC CORP) 22 May 1992 Derwent Accession No. 92-223595/27 , lines 12-22.	1-18

[X] Further documents are listed in the continuation of Box C. [ ] See patent family annex.

*	Special categories of cited documents:	"T"	later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"A"	document defining the general state of the art which is not considered to be part of particular relevance	"X"	document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"E"	earlier document published on or after the international filing date	"Y"	document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"L"	document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	"&"	document member of the same patent family
"O"	document referring to an oral disclosure, use, exhibition or other means		
"P"	document published prior to the international filing date but later than the priority date claimed		

Date of the actual completion of the international search	Date of mailing of the international search report
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Name and mailing address of the ISA/US Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231 Facsimile No. (703) 305-3230	Authorized officer ROBERT SELLERS Telephone No. (703) 308-2351

Form PCT/ISA/210 (second sheet)(July 1992)*

INTERNATIONAL SEARCH REPORT

Inte ..ional application No.
PCT/US94/14550

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US, A, 5,098,780 (NEMUNAITIS ET AL) 24 March 1992, col. 2, lines 48, 50 and 60.	1-18
A	US, A, 5,202,365 (WALLACE) 13 April 1993, col. 4, lines 19-36 and col. 5, lines 26-47.	1-18
A	JP, A, 63-118388, Abstract (AICA KOGYO COCO. LTD.) 23 May 1988, lines 7-10.	1-18
A	US, A, 5,026,794 (HO ET AL.) 25 June 1991, col. 2, lines 53, 54 and 65.	17, 18
A	JP, A, 61-57909 (SHOWA ELECTRIC WIRE) 25 March 1986, Derwent Accession No. 86-116793/18 , see lines 10-12.	12

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US94/14550

BOX II. OBSERVATIONS WHERE UNITY OF INVENTION WAS LACKING
This ISA found multiple inventions as follows:

Group I. Claims 1-14, 17 and 18, drawn to a composition comprising epoxy resin(s), multifunctional (meth)acrylate ester(s), polyurethane poly(meth)acrylate(s) and a polyamine curing agent, classified in class 525, subclass 532.

Group II. Claims 15 and 16, directed to a composition containing epoxy resin(s), multifunctional (meth)acrylate ester(s) and polyurethane poly(meth)acrylate(s), classified in class 525, subclass 532.

The invention of Group I does not form a single general inventive concept based on an intermediate-final product relationship between the intermediate of the composition of Group II and the final product of Group I having an additional polyamine curing agent. The basic chemical structure of the final product is changed due to the reaction between the epoxy resin and polyamine, thereby violating the requirements of Annex B, Part I, section (g) (ii) (A) (1) of the PCT Rule.